

Transient spin dynamics in a single-molecule magnet

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We explore the limitations and validity of semi-classically formulated spin equations of motion. Using a single-molecule magnet as a test model, we employ three qualitatively different approximation schemes. From a microscopic model, we derive a generalized spin equation of motion in which the parameters have a non-local time-dependence. This dynamical equation is simplified to the Landau-Lifshitz-Gilbert equation with i) time-dependent, and ii) time-independent parameters. We show that transient dynamics is essentially non-existing in the latter approximation, while the former breaks down in the regime of strong coupling between the spin and the itinerant electrons. Our analysis suggests that dynamical anisotropy fields strongly depends on how the history of the dynamics is treated, something which has a dramatic effect on the transient dynamics.

I. INTRODUCTION

In the past two decades there has been a rapid progress towards manufacturing single-molecular and single-atomic devices. This has also led towards spin dynamics on pico- and femto-second time scales. There is a large interest in both single-molecular devices and rapid dynamics due to the potential for technical applications, as they pave the way for smaller and faster devices. Except for the technical challenges, approaching smaller systems and faster time-scales question our understanding of dynamics, as quantum and non-equilibrium effects become increasingly important.

Using the intrinsic spin moment of magnetic molecules makes molecular magnets and spintronic devices suitable for logical operations, and serve as good model systems to study fundamental physical phenomena [1–3]. Recent experiments of holmium atoms (Ho) on a manganese oxide-layer (MnO) have shown that one can stabilize single atomic spin moments for long time scales, i.e., hours [4, 5]. This, and other experiments [6–9], open new ways towards realization of single-atom memory devices.

Experimentally it has been shown that one can control the magnetic moment and detect the spin excitations of molecules by electrical current [10–13]. Together with other new methods for probing single-molecule spin states [13–18], future control and read-out of single molecules is possible. Experiments on single magnetic atoms and molecules show distance dependent effects in their exchange [19–22], large anisotropy of individual molecules [23–26], as well as collective spin excitations and Kondo effect [27–30], thus revealing new kinds of physics at the nanoscale. Other experiments revealed the possibility to create quantum mechanical logical gates [31, 32], and to use molecular spintronics for quantum computation [33, 34]. Superconducting spintronics is also a fertile ground for further research [35–39], enhancing the central effects of spintronics devices.

The common approach for describing the dynamics of the magnetization in materials is to employ the phenomenological Landau-Lifshitz-Gilbert (LLG) equation of motion [40]. This has successfully been applied to describe the magnetization dynamics of different materials

[40], and is the commonly used approach when calculating properties and dynamics of for example skyrmions [41] and magnons [42]. In-depth studies have been performed on the Gilbert damping [43–45], which play an important role in the LLG equation. The LLG equation has been extended to take into account temperature, moment of inertia, and stochastic forces [43, 46, 47]. For example, by adding a Langevin term to the equation, one can simulate temperature effects for studies of, e.g., the spin Seebeck effect [48–52].

Due to the pioneering work done in the late 1990s [53–55], ultra-fast demagnetization of materials and ultra-fast spin dynamics have been getting a lot of attention. The physics involved needs insights from both ab-initio methods such as time-dependent density functional theory and atomistic spin dynamics [56]. This has led to further investigation of the LLG equation itself in the ultra-fast regime [46, 47] and magnetic interactions in strongly correlated systems [57].

In this article we focus on the description of the spin dynamics of single-molecule magnets. Methods using quantum master equations [58–61] and stochastic LLG equation [62, 63] have been thoroughly investigated. Another technique, which will be used in this paper, is to derive a spin equation of motion (SEOM) from the spin action defined on the Keldysh contour, considering the nonequilibrium properties of the effective spin moment [44, 64–67]. This provides a general description of the spin dynamics and exchange interactions in the nonequilibrium regime [68–72]. Similar approaches have previously been used in order to understand effects in the stationary limit, such as voltage dependence, geometric phases and chaotic behaviors [44, 73–76]. Here, we study the transient regime, considered through a generalized SEOM where the parameters depend on both time and history.

In this study, we examine the limitations of the LLG equation by comparing three different approximation schemes. First, by making use of the Born-Oppenheimer approximation, one can derive a generalized SEOM where the parameters evolve with time and depends on the full memory of the system. The second approximation scheme is to assume a slowly varying spin, such

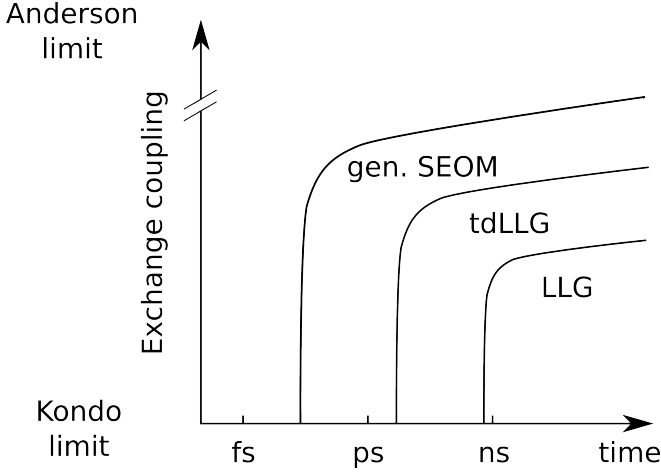


FIG. 1: Diagram showing regimes of validity for the different approximation schemes used in this paper. Going from slower to faster time-scales, and from low to high coupling, one needs to extend the LLG equation to incorporate quantum effects. Here, the generalized SEOM denotes the general approach used in this paper, and tdLLG denotes a LLG equation with time-dependent parameters.

that we can disregard the spin history and retain a LLG equation with time-dependent parameters, henceforth referred to as tdLLG. The third approximation scheme is obtained by considering the parameters of the generalized SEOM in the stationary limit, resulting into a LLG equation with constant parameters. This is the commonly used approach when performing LLG calculations.

Our main result is summarized in Fig. 1. For femtosecond time-scales, the electron dynamics become increasingly important and the Born-Oppenheimer approximation is no longer valid. Therefore, the generalized SEOM is not sufficient and a full quantum mechanical treatment is necessary. This is also true in the Anderson limit, where strong correlations have to be considered for the on-site terms. The generalized SEOM is valid for picosecond time-scales and in systems which can be described by the Kondo model, e.g., a localized spin pertaining to magnetic molecules. Disregarding history, as in tdLLG, one can treat slower dynamics and systems having an exchange coupling less than a few meV. For dynamics in the nanosecond time-scales and weakly coupled, it suffices to use constant parameters in the LLG equation.

The article is organized as follows. In Sec. II, the theoretical background is introduced. This includes a discussion of the LLG equation and a comparison with the derived equation of motion of a spin in nonequilibrium. In Sec. III the simple system studied is described. In Sec. IV the results are presented and discussed, and the article is concluded in Sec. V.

II. THEORY

A. Landau-Lifshitz-Gilbert equation

The phenomenological LLG equation has been successfully applied to a vast range of bulk materials in order to describe their intrinsic magnetism. In its extended form it is defined as [70]

$$\dot{\mathbf{S}} = \mathbf{S} \times (-\gamma \mathbf{B}^{\text{eff}} + \hat{\mathbf{G}}\dot{\mathbf{S}} + \hat{\mathbf{I}}\ddot{\mathbf{S}}), \quad (1)$$

where \mathbf{B}^{eff} , $\hat{\mathbf{G}}$ and $\hat{\mathbf{I}}$ is the effective magnetic field, Gilbert damping and the moment of inertia tensor, respectively. The moment of inertia term, $\hat{\mathbf{I}}$, has here been added in comparison to the conventional LLG equation, as there has been suggestions of its importance to short-time dynamics [43, 70].

If we instead use a microscopic approach [68–70], using non-equilibrium conditions, we can define an effective action of the spin and from that derive generalized SEOM, given by

$$\dot{\mathbf{S}}(t) = \mathbf{S}(t) \times \left(-g\mu_B \mathbf{B}_0^{\text{eff}}(t) + \frac{1}{e} \int \mathbb{J}(t, t') \cdot \mathbf{S}(t') dt' \right). \quad (2)$$

Here, $\mathbf{B}_0^{\text{eff}}(t)$ is the effective magnetic field acting on the spin and $\mathbb{J}(t, t')$ is the dynamical exchange coupling between spins at different times. The effective magnetic field is defined as

$$\mathbf{B}_0^{\text{eff}}(t) = \mathbf{B}^{\text{ext}} + \frac{1}{eg\mu_B} \int \epsilon \mathbf{j}(t, t') dt', \quad (3)$$

where the first term is the external magnetic field and the second term is the internal magnetic field due to the electron flow. In the derivation, we assumed a classical spin of constant length, and ignored quantum fluctuations [77]. While Eq. (1) is an ordinary differential equation, Eq. (2) is an integro-differential equation. Hence, while the former is a simple instant approximation, the latter provides a description based on the whole past evolution of the spin. As we shall see below, this difference has far reaching consequences in the final result.

We retain Eq. (1) from Eq. (2) by assuming that \mathbf{S} is slowly varying with time, $\mathbf{S}(t') \approx \mathbf{S}(t) - (t - t')\dot{\mathbf{S}}(t) + (t - t')^2\ddot{\mathbf{S}}(t)/2$, which leads to

$$\begin{aligned} \frac{1}{e} \int \mathbb{J}(t, t') \cdot \mathbf{S}(t') dt' &\approx \frac{1}{e} \left(\int \mathbb{J}(t, t') dt' \mathbf{S}(t) \right. \\ &\quad \left. - \int \mathbb{J}(t, t')(t - t') dt' \dot{\mathbf{S}}(t) + \int \mathbb{J}(t, t')(t - t')^2 dt' \ddot{\mathbf{S}}(t)/2 \right). \end{aligned} \quad (4)$$

Here, the first term adds a contribution to the effective magnetic field, the second term corresponds to the Gilbert damping and the third term to the moment of

inertia. In the form of the LLG equation we identify the renormalized effective magnetic field

$$\mathbf{B}^{\text{eff}}(t) = \mathbf{B}^{\text{ext}} + \frac{1}{eg\mu_B} \left(\int \boldsymbol{\epsilon} \mathbf{j}(t, t') dt' + \int \mathbb{J}(t, t') dt' \mathbf{S}(t) \right), \quad (5)$$

the damping tensor

$$\hat{\mathbf{G}}(t) = -\frac{1}{e} \int \mathbb{J}(t, t')(t - t') dt', \quad (6)$$

and the moment of inertia term

$$\hat{\mathbf{I}}(t) = \frac{1}{2e} \int \mathbb{J}(t, t')(t - t')^2 dt'. \quad (7)$$

This is still more general than the conventional LLG equation, as the parameters depend on time evolution of the charge and spin background.

B. Exchange coupling

The internal magnetic field due to the electron flow is defined as

$$\boldsymbol{\epsilon} \mathbf{j}(t, t') = ie\epsilon v \theta(t - t') \langle [\mathbf{s}^{(0)}(t), \mathbf{s}(t')] \rangle. \quad (8)$$

Here, v is the exchange integral between the localized and delocalized electrons, $\boldsymbol{\epsilon} = \text{diag}\{\epsilon_\uparrow, \epsilon_\downarrow\}$, the on-site charge is $\mathbf{s}^{(0)} = \sum_{\sigma\sigma'} d_\sigma^\dagger \sigma_{\sigma\sigma'}^0 d_{\sigma'}/2$, where $d_\sigma^\dagger (d_\sigma)$ creates (annihilates) an electron with spin $\sigma = \uparrow, \downarrow$, σ^0 is the identity matrix, and electron spin is denoted $\mathbf{s} = \sum_{\sigma\sigma'} d_\sigma^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} d_{\sigma'}/2$, where $\boldsymbol{\sigma}$ is the vector of Pauli matrices. This two-electron Green function (GF) is approximated by a decoupling into single electron GFs according to

$$\begin{aligned} \boldsymbol{\epsilon} \mathbf{j}(t, t') \approx & iev\theta(t - t') \text{sp} \boldsymbol{\epsilon} \left(\mathbf{G}^<(t', t) \boldsymbol{\sigma} \mathbf{G}^>(t, t') \right. \\ & \left. - \mathbf{G}^>(t', t) \boldsymbol{\sigma} \mathbf{G}^<(t, t') \right), \end{aligned} \quad (9)$$

where $\mathbf{G}^{< / >}(t', t)$ is the lesser/greater matrix GF defined by $\mathbf{G}^<(t, t') = \{i \langle d_{\sigma'}^\dagger(t') d_\sigma(t) \rangle\}_{\sigma\sigma'}$ and $\mathbf{G}^>(t, t') = \{(-i) \langle d_\sigma(t) d_{\sigma'}^\dagger(t') \rangle\}_{\sigma\sigma'}$. In Eq. (9) sp denotes the trace over spin 1/2 space.

The current $\mathbb{J}(t, t') = i2ev^2\theta(t - t') \langle [\mathbf{s}(t), \mathbf{s}(t')] \rangle$ is the electron spin-spin correlation function which mediates the interactions between the localized magnetic moment at times t and t' . Analogously as with the internal magnetic field, we decouple this two-electron GF according to

$$\begin{aligned} \mathbb{J}(t, t') \approx & \frac{ie}{2} v^2 \theta(t - t') \text{sp} \boldsymbol{\sigma} \left(\mathbf{G}^<(t', t) \boldsymbol{\sigma} \mathbf{G}^>(t, t') \right. \\ & \left. - \mathbf{G}^>(t', t) \boldsymbol{\sigma} \mathbf{G}^<(t, t') \right). \end{aligned} \quad (10)$$

This current mediated interaction can be decomposed into an isotropic Heisenberg interaction, J_H , and the

anisotropic Dzyaloshinski-Moriya (DM), \mathbf{D} , and Ising, \mathbb{I} , interactions. This can be seen from the product $\mathbf{S} \cdot \mathbb{J} \cdot \mathbf{S}$, which is the corresponding contribution in the effective spin model [69] to $\mathbf{S}(t) \times \mathbb{J}(t, t') \cdot \mathbf{S}(t')$ in the generalized SEOM [77].

After a little algebra we find that the Heisenberg, Dzyaloshinski-Moriya and Ising interactions can be written as

$$\begin{aligned} J_H(t, t') = & iev^2\theta(t - t') \left(G_0^<(t', t) G_0^>(t, t') \right. \\ & - G_0^>(t', t) G_0^<(t, t') - \mathbf{G}_1^<(t', t) \cdot \mathbf{G}_1^>(t, t') \\ & \left. + \mathbf{G}_1^>(t', t) \cdot \mathbf{G}_1^<(t, t') \right), \end{aligned} \quad (11a)$$

$$\begin{aligned} \mathbf{D}(t, t') = & -ev^2\theta(t - t') \left(G_0^<(t', t) \mathbf{G}_1^>(t, t') \right. \\ & - G_0^>(t', t) \mathbf{G}_1^<(t, t') - \mathbf{G}_1^<(t', t) G_0^>(t, t') \\ & \left. + \mathbf{G}_1^>(t', t) G_0^<(t, t') \right), \end{aligned} \quad (11b)$$

$$\begin{aligned} \mathbb{I}(t, t') = & iev^2\theta(t - t') \left(\mathbf{G}_1^<(t', t) \mathbf{G}_1^>(t, t') \right. \\ & - \mathbf{G}_1^>(t', t) \mathbf{G}_1^<(t, t') + [\mathbf{G}_1^<(t', t) \mathbf{G}_1^>(t, t') \\ & \left. - \mathbf{G}_1^>(t', t) \mathbf{G}_1^<(t, t')]^t \right). \end{aligned} \quad (11c)$$

This leads to that we can partition the exchange interaction in the generalized SEOM into

$$\begin{aligned} \mathbf{S}(t) \times \mathbb{J}(t, t') \cdot \mathbf{S}(t') = & J_H(t, t') \mathbf{S}(t) \times \mathbf{S}(t') \\ & + \mathbf{S}(t) \times \mathbb{I}(t, t') \cdot \mathbf{S}(t') \\ & - \mathbf{S}(t) \times [\mathbf{D}(t, t') \times \mathbf{S}(t')]. \end{aligned} \quad (12)$$

Effectively this corresponds to the Hamiltonian

$$\mathcal{H} = \mathbf{S} \cdot (J_H \mathbf{S} + \mathbb{I} \cdot \mathbf{S} + \mathbf{D} \times \mathbf{S}). \quad (13)$$

In the adiabatic approximation, this gives the damping

$$\mathbf{S} \times \hat{\mathbf{G}} \cdot \dot{\mathbf{S}} = \hat{G}(J_H) \mathbf{S} \times \dot{\mathbf{S}} + \mathbf{S} \times \hat{G}(\mathbb{I}) \cdot \dot{\mathbf{S}} - \mathbf{S} \times (\hat{G}(\mathbf{D}) \times \dot{\mathbf{S}}), \quad (14)$$

and moment of inertia

$$\mathbf{S} \times \hat{\mathbf{I}} \cdot \ddot{\mathbf{S}} = \hat{I}(J_H) \mathbf{S} \times \ddot{\mathbf{S}} + \mathbf{S} \times \hat{I}(\mathbb{I}) \cdot \ddot{\mathbf{S}} - \mathbf{S} \times (\hat{I}(\mathbf{D}) \times \ddot{\mathbf{S}}). \quad (15)$$

It is important to note here, that the exchange coupling mediates both isotropic and anisotropic terms in both the effective magnetic field, the Gilbert damping and the moment of inertia tensor in the framework of the LLG equation. In its general form, this is also mediated in time, since the electronic structure depends on the spin dynamics.

The above treatment incorporates a current driven spin transfer torque, i.e., $\mathbf{S} \times [\mathbf{I}_S \times \mathbf{S}]$, where \mathbf{I}_S is the spin current through the system [40]. This is included in the DM interaction, last term in Eq. (12). The DM interaction can be interpreted as a current through the system and describes a general form of spin current mediated interaction. It is analogous to the spin transfer torque term found in similar treatments of out-of-equilibrium spin systems [78, 79].

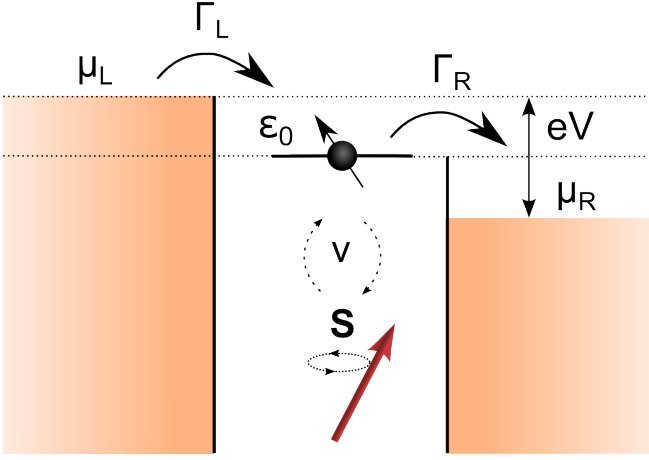


FIG. 2: The system studied in this work consisting of a local magnetic moment coupled to a QD in a tunnel junction between non-magnetic leads.

C. Stationary limit

In the stationary limit, the exchange coupling and the parameters of the equation of motion simplifies further. Ignoring the moment of inertia term, our equation of motion becomes

$$\dot{\mathbf{S}}(t) = \mathbf{S}(t) \times \left(-g\mu_B \mathbf{B}^{\text{eff}} + \hat{\mathbf{G}} \cdot \dot{\mathbf{S}}(t) \right). \quad (16)$$

The electron spin-spin correlation function can be Fourier transformed into energy space

$$\mathbb{J}(\epsilon) = \frac{e}{2} v^2 \int \frac{1}{\omega + \epsilon - \omega' + i\delta} \text{sp} \sigma \left(\mathbf{G}^<(\omega) \sigma \mathbf{G}^>(\omega') - \mathbf{G}^>(\omega) \sigma \mathbf{G}^<(\omega') \right) \frac{d\omega}{2\pi} \frac{d\omega'}{2\pi}, \quad (17)$$

where we used the fact that the GF can be rewritten as $G(t, t') = G(t - t')$ in the stationary limit. This can analogously to the time-dependent case be decomposed into a Heisenberg, Ising and DM term, as done in Ref. [69, 77].

The Gilbert damping can in the stationary limit be derived from [70]

$$\begin{aligned} \hat{\mathbf{G}} &= -\frac{1}{e} \int \mathbb{J}(t, t') (t - t') dt' = -\frac{1}{e} \lim_{\epsilon \rightarrow 0} i \partial_\epsilon \mathbb{J}(\epsilon), \\ &= -\frac{1}{2} v^2 \text{Im} \int \frac{1}{(\omega - \omega' + i\delta)^2} \text{sp} \sigma \left(\mathbf{G}^<(\omega) \sigma \mathbf{G}^>(\omega') - \mathbf{G}^>(\omega) \sigma \mathbf{G}^<(\omega') \right) \frac{d\omega}{2\pi} \frac{d\omega'}{2\pi}. \end{aligned} \quad (18)$$

III. MODEL SYSTEM

Next, we bring the general framework to a specific set-up and we consider a magnetic molecule embedded in a

tunnel junction between metallic leads, see Ref. [77], and Fig. 2 for reference. The magnetic molecule comprises a localized magnetic moment \mathbf{S} coupled via exchange to the highest occupied molecular orbital (HOMO) or lowest unoccupied molecular orbital (LUMO) level, henceforth referred to as the QD level. We define our system Hamiltonian as

$$\mathcal{H} = \mathcal{H}_\chi + \mathcal{H}_T + \mathcal{H}_{\text{QD}} + \mathcal{H}_S. \quad (19)$$

Here,

$$\mathcal{H}_\chi = \sum_{\mathbf{k}\sigma \in \chi} (\varepsilon_{\mathbf{k}\chi} - \mu_\chi) c_{\mathbf{k}\chi\sigma}^\dagger c_{\mathbf{k}\chi\sigma}, \quad (20)$$

is the Hamiltonian for the lead $\chi = L/R$, where $c_{\mathbf{k}\chi\sigma}^\dagger$ ($c_{\mathbf{k}\chi\sigma}$) creates (annihilates) an electron in the lead with energy $\varepsilon_{\mathbf{k}\chi}$, momentum \mathbf{k} and spin $\sigma = \uparrow, \downarrow$. We have introduced the chemical potential μ_χ for the leads and the voltage bias V across the junction defined as $eV = \mu_L - \mu_R$. Each lead has the same temperature T . Tunneling between the leads and the QD level is described by $\mathcal{H}_T = \mathcal{H}_{TL} + \mathcal{H}_{TR}$, where

$$\mathcal{H}_{T\chi} = T_\chi \sum_{\mathbf{k}\sigma \in \chi} c_{\mathbf{k}\chi\sigma}^\dagger d_\sigma + H.c. \quad (21)$$

Using the wide-band limit we can define the tunneling coupling $\Gamma^\chi = 2|T_\chi|^2 \sum_{\mathbf{k} \in \chi} \delta(\omega - \varepsilon_{\mathbf{k}})$ between the lead and the QD. The single-level QD is represented by $\mathcal{H}_{\text{QD}} = \sum_\sigma \varepsilon_\sigma d_\sigma^\dagger d_\sigma$, where d_σ^\dagger (d_σ) creates (annihilates) an electron in the QD with energy $\varepsilon_\sigma = \varepsilon_0 + g\mu_B B \sigma_{\sigma\sigma}^z/2$ and spin σ . We include the Zeeman split due to the external magnetic field $\mathbf{B} = B\hat{\mathbf{z}}$, where g is the gyromagnetic ratio and μ_B the Bohr magneton. The local spin is described by

$$\mathcal{H}_S = -g\mu_B \mathbf{S} \cdot \mathbf{B} - v\mathbf{s} \cdot \mathbf{S}. \quad (22)$$

In the case of non-magnetic lead and vanishing external magnetic field the GF takes the form

$$\mathbf{G}(t, t') = g_0(t, t') - v \oint_C g_0(t, \tau) \langle \mathbf{S}(\tau) \rangle \cdot \sigma g_0(\tau, t') d\tau, \quad (23)$$

where $g_0(t, t')$ is the bare GF for the QD.

IV. RESULTS

In the following we shall refer to Eq. (2) as the generalized SEOM, while the time-dependent LLG equation, Eq. (1), will be referred to as the tdLLG, with parameters given by Eqs. (5)-(7), and constant LLG with parameters given by Eqs. (16) and (18).

The first test is to see if the results of the generalized SEOM can be recreated in different limits by the approximations, e.g., low exchange and tunneling coupling. In

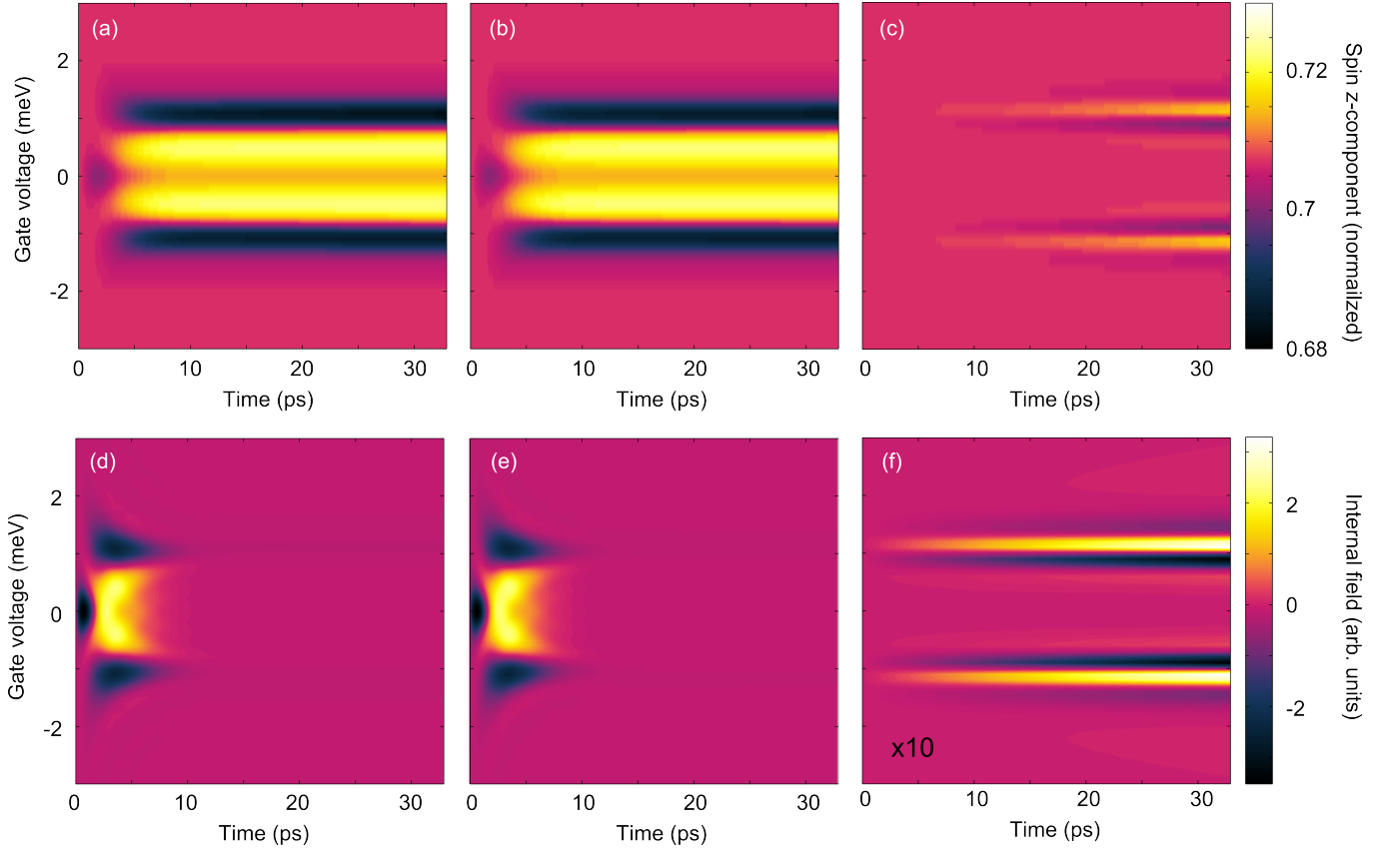


FIG. 3: Evolution of S_z for different gate voltage for (a) the generalized SEOM, (b) the tdLLG solution and (c) the stationary LLG solution. In the bottom row the internal field from the charge flow in the QD, $\int \epsilon \mathbf{j}(t, t') dt'$, is shown for (d) the generalized SEOM, (e) the tdLLG solution and (f) the stationary LLG solution (multiplied by 10). Here, $\Gamma = 0.3$ meV, $v = 0.1$ meV, $V = 2$ meV, $B = 1$ mT and $T = 1$ K.

order to study the transient regime, the system has an abrupt on-set of a voltage bias and the exchange interaction at time t_0 .

We begin by considering the low coupling regime for symmetrically coupled leads, i.e., $v < \Gamma_L = \Gamma_R = \Gamma < 1$ meV. In Fig. 3(a) the solution of the full equation of motion is shown together with the solution of the tdLLG equation, Fig. 3(b), and the solution for the LLG with constant parameters, Fig. 3(c). The plots show the S_z component of the local spin and its evolution due to the sudden on-set. We see that for the generalized SEOM, Fig. 3(a), there are sudden changes in the transient regime and after about 10 picoseconds the system has stabilized and achieved a stationary solution. For the tdLLG equation, Fig. 3(b), it is clear that the generalized SEOM is reproduced. However, in the case of constant parameters, Fig. 3(c), the solution clearly differs from the other two. This can be observed in the internal field, $\int \epsilon \mathbf{j}(t, t') dt'$, shown in Fig. 3(d)-(f) for the three different schemes. The internal field changes rapidly after the on-set, while it increases adiabatically for the stationary solution (do also note the difference of the size of the field by an order of magnitude). Thus, due to the failure to capture the rapid dynamics, the solution for the con-

stant parameters differs from the solution of the dynamic ones. This result shows the importance of treating the dynamic properties of the equation of motion in the picosecond regime and a failure of a stationary treatment in a system far from equilibrium.

Thus far, we have only included damping. Considering the weak coupling regime, i.e., where the exchange coupling v and tunneling coupling Γ is small, higher order terms in the generalized SEOM, such as moment of inertia, will be of importance. Comparing the generalized SEOM, Fig. 4(a), with the tdLLG solution with only damping, Fig. 4(b), and with both damping and inertia, Fig. 4(c), one observe that higher order terms are needed to retain the fast dynamics for the first 2-3 picoseconds. Only using damping, Fig. 4(b), fails to recreate the kink shown in Fig. 4(a), while adding the inertia, Fig. 4(c), captures the effect, although with a small overshoot. Adding even higher order terms is likely providing a solution that converges towards Fig. 4(a). Therefore, we see here that when there is a weak coupling to the magnetic moment and quantum dot, inertia effects will have a larger role in the dynamics as the internal effects start to dominate.

Going to the strong coupling regime, i.e., where the

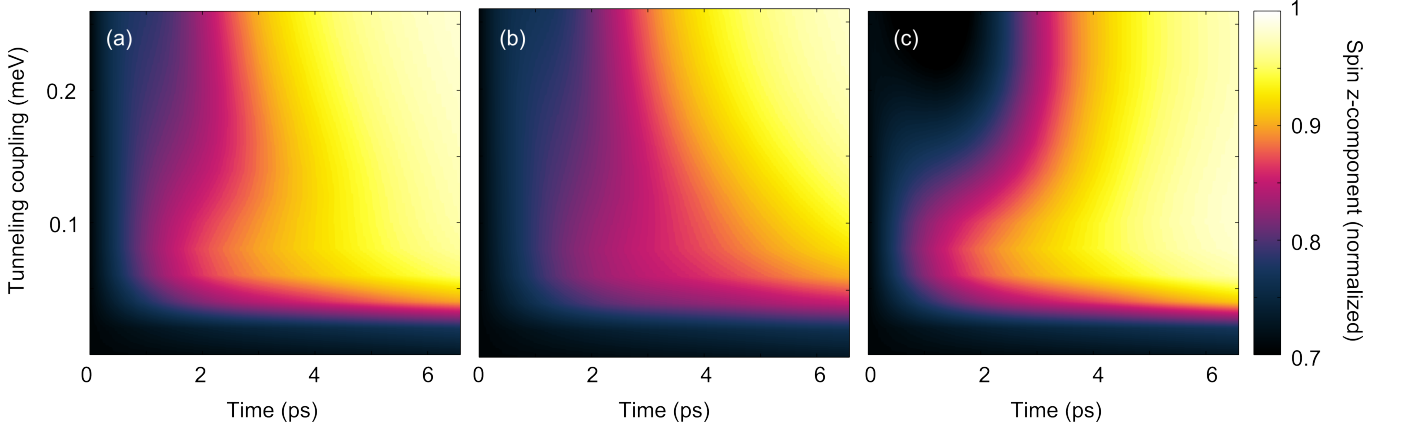


FIG. 4: Evolution of S_z for different tunneling coupling Γ for (a) the generalized SEOM, (b) the tdLLG solution with damping and (c) the tdLLG solution with both damping and moment of inertia. Here, $v = 1.5 \Gamma$, $V = 2$ meV, $B = 1$ T and $T = 1$ K.

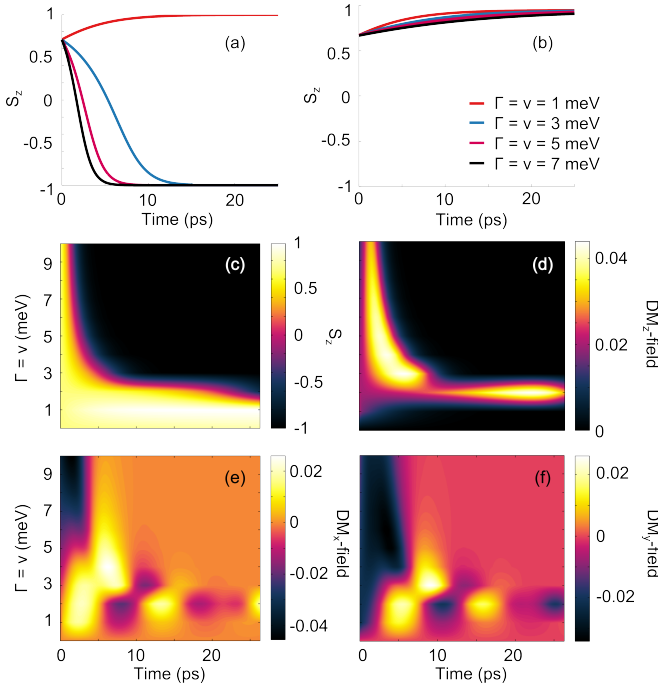


FIG. 5: Evolution of S_z for (a) the generalized SEOM and (b) the tdLLG for different $\Gamma = v$. Contour plot of S_z for the generalized SEOM (c) and the corresponding DM-field in the z-, x- and y-direction in (d), (e) and (f), respectively. Here, there is no applied gate and bias voltage, $B = 1$ T and $T = 1$ K.

exchange coupling v and tunneling coupling Γ are large, we find that even the tdLLG fails to provide an accurate description of the dynamics. In Fig. 5(a)-(b) plots are shown for the evolution of the spin z-component as the interactions are turned on for (a) the generalized SEOM and (b) the tdLLG solution. Here, we apply no gate or bias voltage, only interactions in order to see the effect of the exchange coupling and tunneling coupling on the system. For low exchange coupling and tunneling

coupling, $v = \Gamma < 1$ meV, the solution of both the generalized SEOM, Fig. 5(a), and the tdLLG, Fig. 5(b), are the same. Increasing the coupling parameters gives rise to back-action in the generalized SEOM, Fig. 5(a), which in turn flips the spin moment, something that is not present in the tdLLG solution due to the lack of feedback in the equation. This can be illustrated by the contour plots in Fig. 5(c)-(f), where the generalized SEOM, Fig. 5(c), is shown for different couplings and Fig. 5(d)-(f) shows the corresponding DM-field in the z-, x- and y-direction, respectively. Thus, for the generalized SEOM in the strong coupling regime, the effective fields acting on the spin exert a significantly larger torque on the spin, specially from the Heisenberg interaction and DM-interaction. These non-adiabatic effects cannot be captured by a more simplistic treatment of the equation, as observed in Fig. 5(b). Therefore, the tdLLG solution breaks down in the case of strong coupling, and the stationary state of the system also differs due to the nature of the initial dynamics of the strong-coupled system.

V. CONCLUSION

In conclusion we have compared three different approximation schemes for treating the transient spin dynamics of a magnetic molecule. The results are summarized in Fig. 1. They show that conventional LLG with constant parameters does not capture the fast dynamics in the system, while the tdLLG fails to capture the strongly coupled regime. Therefore, inclusion of the full history in the time-evolution is needed when approaching fast dynamics for exchange couplings above 1 meV. By using a generalized SEOM, we can incorporate the changes in the electronic background, and thereby treat faster dynamics. While our study has been restricted to a single molecule, we believe that our results have implications in larger nanostructures and, hence, the interpretations and validity of spin dynamics using ab initio methods.

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